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The parent program focues on minimization of power budget for the efficient creation of large volume, atmospheric pressure air plasmas, with minimum electron density of 10^{13}cm^{-3} and heavy species translational temperature not exceeding 2000K. Since a thermal plasma with 10^{13}cm^{-3} electron density necessitates a heavy species temperature of approximately 4200 K, the stated program goals inherently define a nonequibibrium plasma. OSU's approach for attaining the program goals is two-pronged based on:
i. Creation of a low translational temperature, highly vibrationally excited environment in which the rates of principal electron loss channels are reduced by orders of magnitude.
ii. Efficient electron production by means of an electron beam and/or application of a short pulse, high E/n electri field.

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"CREATION AND MEASUREMENT OF NONEQUILIBRIUM AIR PLASMAS "

AFOSR Grant #F 49620-00-1-0359

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Final Report

I INTRODUCTION AND BACKGROUND

This final report documents activity and capital equipment expenditures supported by AFOSR supplemental instrumentation grant # F49620-00-1-0359, entitled "Creation and Measurement of Nonequilibrium Air Plasmas." The instrumentation program is aligned with parent AFOSR grant # F49620-97-1-0312, entitled "Plasma Ramparts Using Metastable Molecules." The parent program focuses on minimization of power budget for the efficient creation of large volume, atmospheric pressure air plasmas, with minimum electron density of 10^{13} cm^{-3} and heavy species translational temperature not exceeding 2000K. **Since a thermal plasma with 10^{13} cm^{-3} electron density necessitates a heavy species temperature of approximately 4200 K, the stated program goals inherently define a nonequilibrium plasma.** OSU's approach for attaining the program goals is two-pronged based on:

i. Creation of a low translational temperature, highly vibrationally excited environment in which the rates of principal electron loss channels are reduced by orders of magnitude.

ii. Efficient electron production by means of an electron beam and/or application of a short pulse, high E/n electric field.

In particular, supplemental equipment funding was requested for the following items:

i. A GEN IV Intensified CCD/Optical Multichannel Analyzer (OMA) system for use as the detector for pulsed Raman scattering diagnostics and for a novel spectrally filtered rotational Raman/Thomson scattering apparatus, currently under development at OSU. The technique employs an atomic rubidium vapor filter as a means to greatly attenuate interferences from Rayleigh/Mie scattering, while transmitting the incoherent rotational Raman/Thomson scattering signal.

ii. Dedicated emission - mode Fourier Transform - Infra Red spectrometer, for determination of CO vibrational distribution functions and translational/rotational temperature of the nonequilibrium plasma environment.

The creation of large volume, low temperature, atmospheric pressure air plasmas, with even modest ionization fraction (order 10^{-6}), constitutes an enormous challenge. Fundamentally, at standard sea level temperature (298K) and pressure (1 atm.) conditions, the power required to sustain a steady-state electron number density of 10^{13} cm^{-3} is estimated to be as high as on the order of 10 GW/m^3, assuming that the predominant loss mechanism is three-body attachment of

free electrons to oxygen. Fortunately, the rate of electron attachment to O_2 is known to rapidly decrease with increasing temperature. However, even if the loss rate from this process could be made negligible, the process of dissociative recombination (for example, $e^- + O_2^- \rightarrow O^- + O$) would lead to power requirements on the order of 1 GW/m^3. Additionally, it must be noted that this rather formidable requirement assumes 100% coupling of the applied power to the electrons. **It is clear, therefore, that the development of practical air plasma-based engineering devices will require <u>both</u> efficient mechanisms for electron production <u>and</u> novel techniques for reduction of electron loss rates.**

These considerations have led to the assembly of the OSU/Princeton "definitive experiment," which is centered around the concept of using an electron beam as an efficient volume ionization source, combined with what is known as CO laser "optical pumping". Work performed at OSU under the parent program (see bib 3) has established that optical pumping can be used to vibrationally excite all diatomic species in an atmospheric pressure air plasma to a vibrational temperature of order 2,000 K (or greater) while maintaining low (order ~400 – 500 K) translational/rotational temperature. This suggested the hypothesis, confirmed recently (bib 7,9,10,14) that vibrational excitation, provided by optical pumping, could be used to reduce the overall electron removal rate in e-beam sustained, atmospheric pressure air plasmas, resulting in a significantly reduced power budget.

II. UTILIZATION OF EQUIPMENT

In this section we briefly summarize measurements obtained (and planned in the near future) using the equipment items purchased with funds from this grant. Much more detail can be found in the publications cited in the bibliography and in the FY 2001 parent grant progress report, which covers the period Sept. 1, 2000 – Aug. 31, 2001.

<u>FT-IR Measurments</u>

Figure 1, below, shows a simplified schematic illustrating the essential features of the OSU/Princeton definitive experiment, incorporating the FT – IR spectrometer and the OMA/Pulsed Raman system. The CO pump laser pump and Raman/Thomson scattering diagnostics (probe) laser are collinearly directed into the plasma cell perpendicular to the propagation direction of the e-beam. Electron density is determined by microwave attenuation employing a pair of wave guides, which for clarity, are not included in Fig. 1.

The FTIR spectrometer is used in emission mode to obtain the vibrational distribution function (VDF) of CO and/or rotational temperature of the optically pumped/e-beam generated plasma. Fig. 2 shows a typical rotationally resolved CO fundamental R-branch emission spectrum from which a heavy species temperature is extracted by means of equilibrium Boltzmann statistics. In the case illustrated, the optically pumped plasma was created under conditions identical to that used in bibs 9,10,14 which demonstrated that vibrational excitation resulted in a minimum three order of magnitude reduction in the net rate of electron attachment to oxygen:

$$O_2(v)+e^- +M \leftrightarrow O_2^-(v)+M \qquad (1)$$

and one order of magnitude reduction in the rate of dissociative recombination

$$O_2^+(v)+e^- \rightarrow O+O. \qquad (2)$$

The measured rotational temperature is 581 K, which is not, in it self, sufficiently high to account for the measured reduction in the recombination rate.

The FT – IR system has also been used for CO VDF and rotational temperature measurements performed as part of a related parent grant project to develop a new CO planar laser induced fluorescence imaging diagnostic (Bibs 8,9), fundamental optical pumping and RF energy transfer studies (bibs 1,2,5,6,13,15), plasma aerodynamic studies (bib 12), studies of dusty plasmas (bib 4), and isotope separation studies.

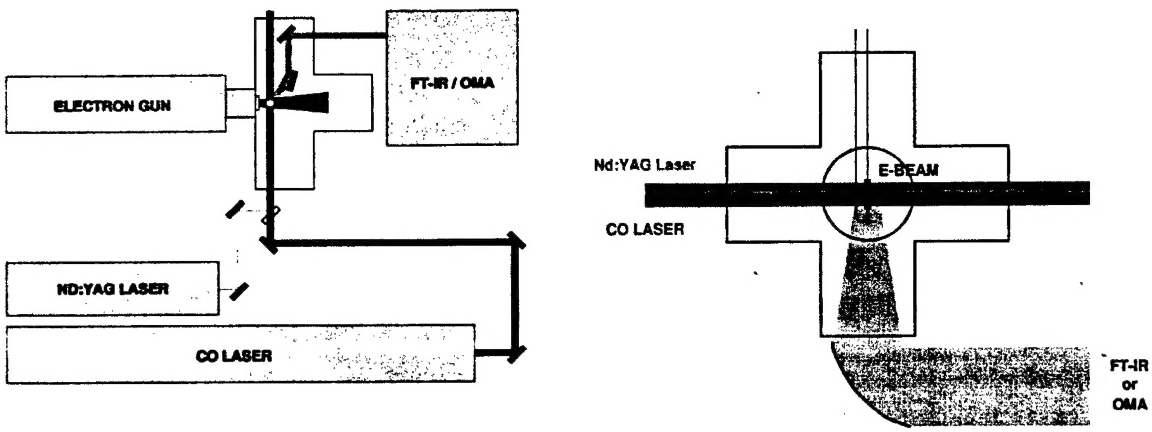

Figure 1.
Schematic of electron beam / optical pumping experiment (left)
and blow up of optical absorption cell (right).

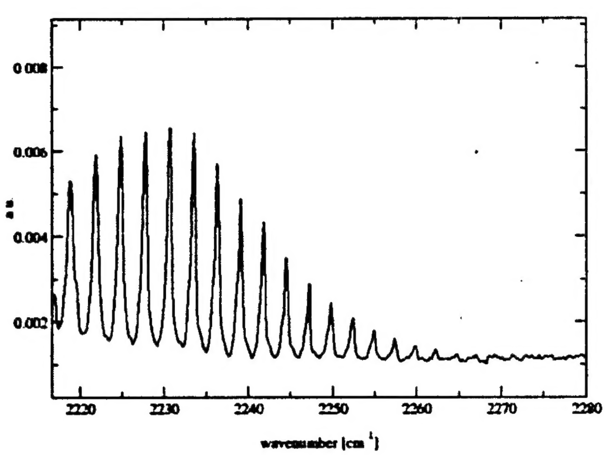

Figure 2
Typical rotationally resoloved FT – IR emission spectrum used to infer heavy species temperature in OSU/Princeton definitive experiment.

Raman Scattering Measurments

Raman scattering measurements have been performed to verify that optical pumping can be used to produce substantial vibrational nonequilibrium in high (up to atmospheric) pressure air (bibs). Figure 3 shows typical vibrational Raman spectra obtained from a mixture of 40 torr CO, 120 torr O_2, and 580 torr N_2. The top spectrum shows five N_2 vibrational levels, v=0-4, with corresponding vibrational temperature, defined by consideration of the populations of levels 0 and 1 only, of $T_{v,N2}$ =2100±70 K. The middle spectrum shows the v=0-6 bands of CO, with $T_{v,CO}$=3050±200 K. The bottom spectrum contains vibrational states of O_2, which shows significant population in levels v=0-6, with T_v=2200±150 K. Note that the full vibrational distributions of CO and O_2 are strongly non-Boltzmann and cannot be truly characterized by a single vibrational temperature.

The data of Fig. 3, (as well as other spectra given in bib #) represent, to our knowledge, the first steady-state optical pumping experiments performed in high pressure (up to 1 atm) mixtures of diatomic gases. In particular, it is clear from the data of Fig. 3 that a relatively low intensity (~100 W/cm^2) CO laser can be used to produce steady state optical pumping in atmospheric pressure air with effective vibrational temperatures exceeding 2000 K for all three major diatomic species, while maintaining relatively low (~ 400 – 500 K) translational/rotational temperature.

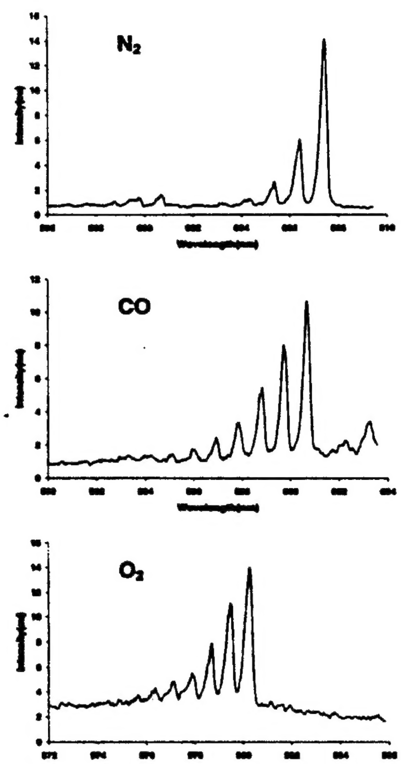

Figure 3
Experimental Raman spectrum of optically pumped 580/120/40 torr mixture of $N_2/O_2/CO$.

Spectrally Filtered Rotational Raman/Thomson Scattering

Recent advances in solid state laser technology have enabled a variety of new optical diagnostic techniques based on the use of atomic/molecular vapor filters as narrow bandwidth "notch" filters and/or as spectral discriminators. During the past year, a filtered rotational Raman/Thomson scattering diagnostic has been developed which employs a rubidium vapor filter in combination with a diode laser injection- seeded, narrow spectral bandwidth titanium:sapphire laser at 780 nm. The particular emphasis is to use the rubidium vapor filter as an ultra narrow band (~ 1-2 GHz) pre-absorption filter in order to attenuate elastic and/or Rayleigh scattering, while transmitting low wave number inelastic scattering diagnostics, such as pure rotational Raman and Thomson scattering. With this diagnostic, the spatially resolved measurement of both gas temperature and electron density in our air plasmas will be possible.

As an example of the capabilities of the system, Figs. 4 and 5 show pure rotational Raman spectra of N_2 obtained at 500 torr pressure and room temperature, obtained without (Fig. 4) and with (Fig. 5) incorporation of the rubidium filter (from bib 8). Note that the intensity axes of the spectra are normalized to the approximate peak intensity of the maximum detected Rayleigh/Mie scattering signal. It can be seen from Fig. 4b (right), which is a factor 500 blow-up of the spectrum displayed in Fig. 4a, that the maximum detected pure rotational Raman signal is ~ 0.08% of the detected unfiltered Rayleigh/Mie signal. It is this intense, interfering "quasi-elastic" scattering which has traditionally prohibited Thomson scattering measurements in high pressure gases. However, as can be seen in Fig. 5, employing a vapor filter greatly reduces this interfering signal. In Fig. 5, the detected pure rotational Raman intensity is a factor of ~ 50 times greater than that of the filtered Rayleigh signal, indicating that the filter has attenuated the Rayleigh scattering by a factor of ~ 50,000.

We plan to incorporate this diagnostic into the OSU/Princeton electron beam experiment during the remaining portion of the MURI program. The goal will be to perform accurate (+/- 20 K or better) spatially and temporally resolved rotational temperature measurements, as well as spatially and temporally resolved electron density, and, perhaps, electron temperature measurements.

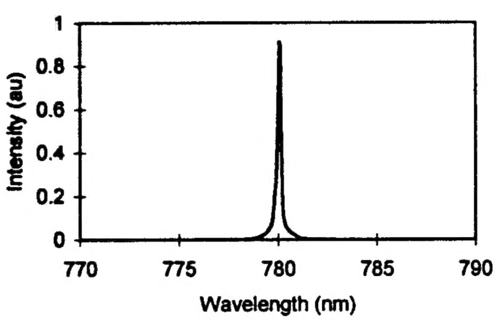

Fig 4a: Pure rotational Raman spectrum of 500 torr N_2 at room temperature. No vapor filter employed.

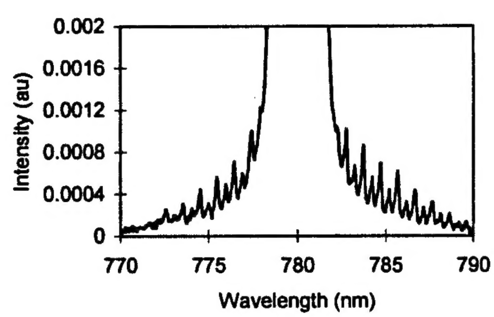

Fig 4b: 500 X blow up of Fig. 4a

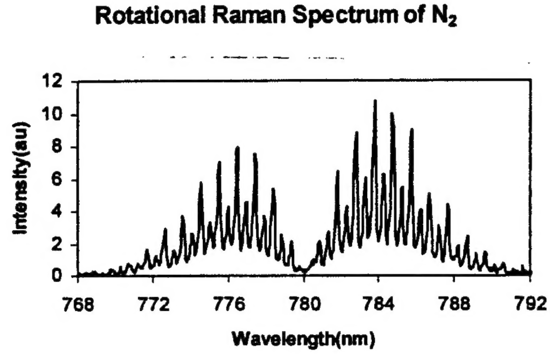

Figure 5
Pure rotational Raman spectrum of 500 torr of N_2 at room temperature employing rubidium vapor filter.

III STUDENTS SUPPORTED

The instrumentation has been and is currently being used to directly support the thesis research of six Ph.D students, Robert Leiweke, Wonchul Lee, Kraig Frederickson, Tai Ahn, Katherine Essenhigh, and Alan White, one Masters student, Jared Lilley, and one undergraduate student, Matt Buoni.

IV BIBLIOGRAPHY OF PAPERS EMPLOYING INSTRUMENTATION

Journal papers with parent grant support

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4. J. L. Lilly, and V. V. Subramaniam, "A Cold Thermionically-Emitting Dusty Air Plasma Formed by Radiative Heating of Graphite Particulates", accepted for publication in IEEE Trans. Plasma Sci.

5. E. Plönjes, P. Palm, J.W. Rich, and I.V. Adamovich, "Electron-Mediated Vibration-Eelectronic (V-E) Energy Transfer in Optically Pumped Plasmas", submitted to Journal of Chemical Physics, August 2001.

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